



PATENT ABSTRACTS OF JAPAN

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FUJII YASUHIRO(54) **REGENERATING METHOD FOR SOOT
PARTICLE AND NITROGEN OXIDE
ELIMINATING DEVICE, AND ELIMINATING
METHOD FOR SOOT PARTICLE AND
NITROGEN OXIDE**

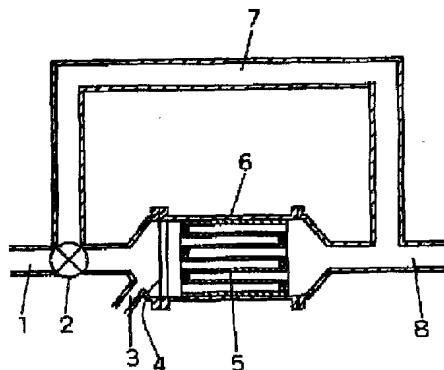
(57) Abstract:

PURPOSE: To maintain high purging ability for a long time by burning dust captured in a filter, forming the predetermined reducing atmosphere by using the combustion, and reducing absorbed nitrogen oxide.

CONSTITUTION: When exhaust gas is passed through a filter 5, an elimination of dust and nitrogen oxide is carried out. When a back pressure increased because of accumulation of dust or absorbing ability for nitrogen oxide exceeds the preset value, a valve 2 is switched, and inflow of the exhaust gas to the filter 5 is stopped. Then, air is fed to the filter 5 by means of a pump, and the air and the filter 5 are heated by means of a heater 4. When combustion of the dust is

started following the heating, oxygen around the filter 5 is consumed. At this time, an air feeding quantity is regulated so as to make the vicinity of the filter 5 a reducing atmosphere, and consequently, nitrogen oxide is reduced on a catalyst. In this way, purging ability can be maintained for a long time.

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CLAIMS

[Claim(s)]

[Claim 1] By heating a filter which performs prehension of soot dust, and absorption of nitrogen oxides, burning soot dust caught by said filter, making predetermined reducing atmosphere using the combustion, and returning said absorbed nitrogen oxides, A regeneration method of a stripper of soot dust and nitrogen oxides reproducing said filter.

[Claim 2] With a filter which has a prehension function of soot dust, and an oxidation absorption function of nitrogen oxides. While burning soot dust caught by said filter by heating said filter and making said neighborhood of a filter into hypoxia atmosphere before it purifies soot dust and nitrogen oxides and absorptance of nitrogen oxides or capturing capacity power of soot dust declines, A removing method of soot dust and nitrogen oxides completing combustion of soot dust which remains in said filter, and reproducing said filter after returning nitrogen oxides absorbed by said filter and recovering absorptance of nitrogen oxides.

[Claim 3] With a filter which has a prehension function of soot dust, and an oxidation absorption function of nitrogen oxides. Purify soot dust and nitrogen oxides, and said filter is heated before absorptance of nitrogen oxides or capturing capacity power of soot dust declines, And while performing combustion of soot dust and steam reforming of soot dust which were caught by said filter by adding a steam and making said neighborhood of a filter into hypoxia atmosphere, A removing method of soot dust and nitrogen oxides completing combustion of soot dust which remains in said filter, and reproducing said filter after returning nitrogen oxides absorbed by said filter and recovering absorptance of nitrogen oxides.

[Claim 4] When an oxygen supply part is provided downstream from a filter, a catalyst which has an oxidation function is installed downstream from said oxygen supply part and an unburnt ingredient is discharged from said filter, A removing method of soot dust according to claim 2 or 3 and nitrogen oxides introducing gas which contains oxygen from said oxygen supply part, and removing said unburnt ingredient according to said oxidation catalyst.

[Claim 5] A removing method of soot dust according to claim 4 and nitrogen oxides using for heating at the time of filter regeneration reaction fever generated in an oxidation catalyst.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the regeneration method of the stripper of soot dust and nitrogen oxides available when, removing simultaneously the soot dust and nitrogen oxides which are discharged from a diesel engine etc. for example, and the removing method of soot dust and nitrogen oxides.

[0002]

[Description of the Prior Art] In recent years, the air pollution by nitrogen oxides and soot dust in a metropolitan region poses a problem, and an immediate measure is desired. If these are roughly divided, it will be divided into what is discharged from the stationary source of a factory etc., and the thing discharged from the mobile sources of a car etc., but especially the nitrogen oxides and soot dust from a diesel rolling stock pose a big problem triggered by the difficulty of the yield and purification.

[0003] In order to solve the aforementioned problem in a diesel rolling stock, engine improvement of the improvement of an air-supply-and-exhaust system by an improvement of a combustion chamber, an improvement of a fuel injection system, supercharge, etc., exhaust gas recirculation, etc. is considered, and nitrogen oxides and soot dust are being reduced. In order to solve an aforementioned problem, the improvement of fuel or a lubricating oil is also tried.

[0004] Development of the removal of an inflammable organic component (SOF) by an oxidation catalyst, prehension of the soot dust by a diesel particulate filter (DPF), a NO_x reduction catalyst, etc. is furthered as post-processing art of exhaust gas.

[0005]

[Problem(s) to be Solved by the Invention] However, in a diesel power plant, discharge control of nitrogen oxides and discharge control of soot dust have a relation of a trade-off, and there is a limit in those reduction effects only by engine improvement. The improvement of fuel or a lubricating oil also has many the problems and technical technical problems of cost.

[0006] In removal of SOF by the oxidation catalyst which is post-processing art, technical problems, such as improvement in low-temperature activity and control of sulfate generation at an elevated temperature, occur, and nitrogen oxides also have the problem that it cannot control. Although the crack initiation by the temperature gradient at the time of reproduction is a problem and DPF is being solved by improvement of a regeneration method, nitrogen oxides cannot be controlled in this method. For this reason, although

development of the NO_x reduction catalyst which operates under hyperoxia conditions like diesel-rolling-stock exhaust gas is furthered, it is still a research stage from points, such as shortage of cleaning capacity and the necessity for a reducing agent.

[0007] Thus, the conventional post-processing art had many problems, and it had the technical problem were difficult that any method removes nitrogen oxides and soot dust simultaneously substantially.

[0008] This invention can remove substantially simultaneously the soot dust and nitrogen oxides which are discharged from a combustion engine in view of such a technical problem of a Prior art, And it aims at providing the regeneration method of the stripper of soot dust and nitrogen oxides which can maintain high removal ability over a long period of time compared with the former more, and the removing method of soot dust and nitrogen oxides, without using special reducing agents, such as hydrocarbon.

[0009]

[Means for Solving the Problem] This invention of claim 1 heats a filter which performs prehension of soot dust, and absorption of nitrogen oxides, It is a regeneration method of a stripper of soot dust and nitrogen oxides which reproduces said filter by burning soot dust caught by said filter, making predetermined reducing atmosphere using the combustion, and returning said absorbed nitrogen oxides.

[0010] This invention of claim 2 with a filter which has a prehension function of soot dust, and an oxidation absorption function of nitrogen oxides. While burning soot dust caught by filter by heating a filter and making the neighborhood of a filter into hypoxia atmosphere before it purifies soot dust and nitrogen oxides and absorptance or soot dust capturing capacity power of nitrogen oxides declines, After returning nitrogen oxides absorbed by filter and recovering absorptance of nitrogen oxides, it is a removing method of soot dust and nitrogen oxides which completes combustion of soot dust which remains in a filter, and reproduces a filter.

[0011] While this invention of claim 3 adds a steam at the time of filter regeneration, performs combustion of soot dust and steam reforming of soot dust which were caught by filter, for example and makes the neighborhood of a filter hypoxia atmosphere, After making nitrogen oxides absorbed by filter return and recovering absorptance of nitrogen oxides, it is a removing method of soot dust and nitrogen oxides which completes combustion of soot dust which remains in a filter, and reproduces a filter.

[0012] Furthermore, when an oxygen supply part is provided downstream from the above-mentioned filter, a catalyst which has an oxidation function is installed downstream from an oxygen supply part and an unburnt ingredient is discharged from a filter, this invention of claim 4, It is a removing method of soot dust and nitrogen oxides which introduces gas which contains oxygen from an oxygen supply part, and removes an unburnt ingredient according to an oxidation catalyst.

[0013] This invention of claim 5 is a removing method of soot dust and nitrogen oxides which uses for heating at the time of filter regeneration reaction fever generated in the above-mentioned oxidation catalyst.

[0014]

[Function] In this invention, the filter for ceramic monolithic wall style type particulates is mentioned, for example as an example of the filter used. This filter includes the catalyst which has a function which oxidizes nitric oxide to nitrogen dioxide under hyperoxia conditions, a function which absorbs nitrogen dioxide, and the function to return the nitrogen oxides which ****ed and were desorbed from the nitrogen oxides which the oxygen density absorbed in the atmosphere below a stoichiometric ratio to combustion of an unburnt ingredient further. As an example of a catalyst used by this invention, the catalyst which supported alkaline-

earth metals, or a rare earth metal oxide and the precious metals to alumina is mentioned. Even nitrogen dioxide can oxidize nitric oxide by the oxidation of the bottom precious metals of a hyperoxia condition, and this catalyst can be made to absorb as a nitrate with metal, such as Ba and La, further. It can oxidize an unburnt ingredient while returning the nitrogen oxides which ****ed and were desorbed from the nitrogen oxides which were being absorbed to nitrogen, if this catalyst heats [an oxygen density] in the atmosphere below a stoichiometric ratio to combustion of an unburnt ingredient. It is thought that reduction of nitrogen oxides and combustion of an unburnt ingredient are the same mechanisms as a three way component catalyst. The multiple oxide containing Y, Ba, and Cu and the multiple oxide containing Mn and Zr may be used as a nitrogen oxide absorbent of this catalyst.

[0015]The regeneration method of the soot dust of this invention and the stripper of nitrogen oxides and the simultaneous removing method of soot dust and nitrogen oxides are explained collectively, describing hereafter the composition and operation of a purge which are shown in drawing 1.

[0016]Usually, exhaust gas is circulated in the filter 5 and removal of soot dust and removal of nitrogen oxides are performed by the above-mentioned principle. When constant value with the absorptance of the back pressure which increases by deposition of soot dust, or nitrogen oxides is exceeded, the valve 2 located in the filter 5 upper stream is switched, and the inflow of the exhaust gas to the filter 5 is suspended. On the other hand, air is supplied to the filter 5 with a pump, and the heater 4 performs heating of air and the filter 5. Although the combustion temperature of soot dust is usually 500-600 **, combustion at low temperature is attained by operation of the catalyst included in the filter 5. although the combustion temperature of the soot dust at the time of using a catalyst is based also on a controlled atmosphere and the description of soot dust -- 300-500 -- it becomes tens of **. If combustion of soot dust starts with heating, about five-filter oxygen will be consumed. Air supply is adjusted so that about five filter may become reducing atmosphere at this time. By this, desorption of the absorbed nitrogen oxides and reduction of the nitrogen oxides by partial oxidation output take place on a catalyst. When desorption of the absorbed nitrogen oxides is fully performed, combustion of the increase of air supply and the soot dust which remains in the filter 5 without burning is completed. In this way, after reproducing the soot dust capturing capacity power of the filter 5, and the absorptance of nitrogen oxides, cleaning capacity is maintainable over a long period of time by purifying exhaust gas again.

[0017]Like a diesel rolling stock, when there are also many sulfur oxides in exhaust gas, a sulfur oxide is also absorbed by the catalyst, but. Since it will **** if the absorbed sulfur oxide is also exposed to an elevated temperature under reducing atmosphere, the fall of nitrogen-oxides absorptance can be controlled by burning the soot dust under reducing atmosphere at an elevated temperature further.

[0018]If a filter is installed in parallel and another filter purifies exhaust gas during filter regeneration of one of the two, purification of exhaust gas can be performed continuously substantially.

[0019]Although here described the method by an electric heater as the filter heating method, it is also possible to carry out direct heating of the soot dust with microwave.

[0020]With a strong substance of reducing power like [if a steam is added at the time of filter regeneration] the carbon monoxide which the oxygen tension near the filter could be reduced more easily and was further generated by steam reforming of soot dust, or hydrogen which burns easily. The conversion efficiency to nitrogen of the absorbed nitrogen oxides can be raised. In this case, it is desirable to make a filter contain the catalyst which promotes steam reforming like platinum or nickel. When desorption of the absorbed

nitrogen oxides is completed, the operation which removes the soot dust which remained on the filter, and the carbon which deposited by steam reforming by combustion is the same as that of the method shown above.

[0021]Although the technical problem that it is generated by partial oxidation output occurs at the time of the reduction to nitrogen of the absorbed nitrogen oxides, This is solvable by providing an oxygen supply part downstream from a filter, installing the catalyst which has an oxidation function downstream from an oxygen supply part, introducing the gas which contains oxygen from an oxygen supply part, and oxidizing an unburnt ingredient according to an oxidation catalyst.

[0022]In the way an oxidation catalyst removes an unburnt ingredient, the reaction fever generated at the time of unburnt ingredient oxidation can be used for heating of a filter by installing an oxidation catalyst near the filter.

[0023]

[Example]Hereafter, the example of this invention is described, referring to drawings.

[0024]The regeneration method of the stripper of the soot dust and nitrogen oxides concerning other inventions is also explained simultaneously, describing one typical example of the removing method of soot dust and nitrogen oxides concerning this invention using drawing 1. Here, the stripper of soot dust and nitrogen oxides corresponds to the filter 5 of this example.

[0025]in drawing 1 -- 1 -- an exhaust gas introducing pipe and 2 -- as for a filter and 6, an air supply part and 4 are [a bypass and 8] exhaust gas discharge pipes a filter holding container and 7 a heater and 5 a valve and 3.

[0026]A below concrete example is explained in more detail.

[0027]A total of 300 g of catalysts which contain alumina, a lanthanum trioxide, barium oxide, cerium oxide, and the precious metals in the exhaust gas filter of the quality of mullite of the capacity of $1 > 3$ l. of < examples was supported. Making the lanthanum trioxide in a catalyst into 50wt%, the precious metals supported a total of Pt and 5 g/l of Rh(s).

[0028]This filter 5 was installed in the exhaust air part of a direct injection diesel engine with a displacement of 2400 cc, the valve 2 was formed in the upstream part of the filter 5, and the bypass 7 which can discharge exhaust gas as it is was installed. The heater 4 was formed in filter 5 front face, and the air supply part 3 was formed between the heater 4 and the valve 2. Space velocity was made into 60000h^{-1} , and emission gas was analyzed by the chemiluminescence type NOx analyzer, the hydrocarbon analyzer, and the gas chromatograph.

[0029]The presentations of filter 5 entrance gas at this time were about 500 ppm of nitric oxide, the sulfur dioxide of about 100 ppm, the amount of total hydrocarbon of about 500 ppm (methane conversion), about 4% of carbon dioxide, about 15% of oxygen, and about 10% of water.

[0030]Nitric oxide could carry out the extraction ratio about 90% at the time of an experiment start, and the extraction ratio fell with time. When the extraction ratio cut 30%, while passing the exhaust gas which circulates the filter 5 by the valve 2 to the bypass 7 and carrying out about 1 l/min supply of the air from the air supply part 3 further, heating of the heater 4 was started. When combustion of soot dust started, the discharged nitrogen oxide concentration became high temporarily, but it fell immediately. After the temperature of the filter 5 became about 650 ** and was fully desorbed from the sulfur compound, the amount of air supply which circulates the filter 5 was raised, and combustion of the soot dust accumulated in

the filter 5 was completed. Usually, it was 33% when the mean clearance rate of nitrogen oxides was calculated from the quantity of the discharge nitrogen oxides at the time of operation, and the quantity of the nitrogen oxides discharged at the time of filter 5 reproduction.

[0031]Although air was used for reproduction here, it is also possible to use a part of exhaust gas which is a hypoxia partial pressure as resurgent gas. In this case, more efficient reproduction is possible if the combustion temperature of the exhaust gas at the time of reproduction is raised by throttling using an inhalation-of-air diaphragm or an exhaust air diaphragm.

[0032]The catalyst of the filter 5 used in <Example 2> example 1 was made to contain nickel oxide. It was made for the weight of nickel oxide to be 10wt% into a catalyst.

[0033]First, like Example 1, perform absorption of nitrogen oxides, and prehension of soot dust, and the air which contained 50vol% of the steam from the air supply part 3 at the time of reproduction is circulated by about 1 l/min, When the temperature of the filter 5 became 650 **, supply of the steam was suspended, only air was circulated, and combustion of soot dust was completed like Example 1. On this condition, from the quantity of the discharge nitrogen oxides at the time of operation, and the quantity of the nitrogen oxides by which it was generated at the time of filter 5 reproduction, when the mean clearance rate of nitrogen oxides was searched for, compared with the case of 38% and Example 1, the nitrogen-oxides extraction ratio usually improved.

[0034]From the above result, by adding a steam in a filter at the time of reproduction showed that the reduction to nitrogen of the absorbed nitrogen oxides was promoted.

[0035]A total of 100 g of catalysts which contain alumina, Pt and Rh, and cerium oxide in the honeycomb of the cordierite of the capacity of 3> 1 l. of < examples was made to support, and the oxidation catalyst 16 was prepared.

[0036]As shown in (drawing 2), the air supply part 15 for unburnt ingredient purification was formed downstream from the filter 13, the oxidation catalyst 16 was installed downstream from the air supply part 15 for unburnt ingredient purification, and the heater 17 which heats the oxidation catalyst 16 was formed.

[0037]First, absorption of nitrogen oxides and prehension of soot dust were performed like Example 1. After that, at the time of reproduction, air was supplied by 1 l/min from the air supply part 15 for unburnt ingredient purification, and the oxidation catalyst 16 was heated at 450 ** with the heater 17. Reproduction was performed by the same method as Example 1, when reproduction was completed, it suspended introduction of the air from the air supply part 15 for unburnt ingredient purification, and heating of the oxidation catalyst 16 also suspended it. When the hydrocarbon concentration of the lower stream of the oxidation catalyst 16 was measured by the hydrocarbon analyzer and it compared with the case of Example 1 at the time of reproduction, it became 17% when the amount of total hydrocarbon discharged at the time of reproduction is not installing the oxidation catalyst 16, and was decreasing greatly.

[0038]As a result, it turned out that the quantity of the unburnt ingredient generated at the time of reproduction can be greatly reduced according to an oxidation catalyst.

[0039]The ***** cordierite honeycomb was created for the central part in the form surrounding [as shown in <Example 4> (drawing 3)] the periphery of the filter holding container 25. Honeycomb capacity was 1 l. By supporting a catalyst like Example 3 to this honeycomb, the oxidation catalyst 27 was prepared and the filter holding container 25 was installed with the wrap form.

[0040]On the other hand, the emission gas from the filter 24 changed the channel so that the oxidation

catalyst 27 might be passed, and it formed the air supply part 26 for unburnt ingredient purification between the filter 24 and the oxidation catalyst 27. The heater 28 was formed so that the periphery of an oxidation catalyst holding container could be heated.

[0041]First, absorption of nitrogen oxides and prehension of soot dust were performed like Example 1. After that, at the time of reproduction, air was supplied by 1 l/min from the air supply part 26 for unburnt ingredient purification, and the oxidation catalyst 27 was heated at 450 **. Reproduction of the filter 24 was performed by the same method as Example 1, when reproduction was completed, it suspended introduction of the air of the air supply part 26 for unburnt ingredient purification, and heating of the oxidation catalyst 27 also suspended it. When the time of the time of the end of desorption of a sulfur compound was measured in the meantime, it turned out that time is shortened by 5 by about 3/in the case of Example 1.

[0042]From the above result, by installing this oxidation catalyst near the filter showed that reaction fever could be used for heating of a filter and a filter could be reproduced efficiently by the way an oxidation catalyst removes an unburnt ingredient.

[0043]Although the above-mentioned example explained the case where a filter was reproduced by completing combustion of the soot dust which reproduced the absorptance of the nitrogen oxides of a filter and was caught by the filter after that by returning the nitrogen oxides absorbed by the filter 5, If the capturing capacity power of the soot dust of a filter is recovered to some extent by combustion of soot dust performed under hypoxia atmosphere when returning the nitrogen oxides absorbed by not only this but the filter, after reproducing the absorptance of nitrogen oxides, it is not necessary to perform combustion of the still more above soot dust.

[0044]

[Effect of the Invention]This invention has the strong point in which high cleaning capacity can be further maintained over a long period of time compared with the former, without using special reducing agents, such as hydrocarbon, for example so that clearly from the place described above.

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TECHNICAL FIELD

[Industrial Application] This invention relates to the regeneration method of the stripper of soot dust and nitrogen oxides available when, removing simultaneously the soot dust and nitrogen oxides which are discharged from a diesel engine etc. for example, and the removing method of soot dust and nitrogen oxides.

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PRIOR ART

[Description of the Prior Art] In recent years, the air pollution by nitrogen oxides and soot dust in a metropolitan region poses a problem, and an immediate measure is desired. If these are roughly divided, it will be divided into what is discharged from the stationary source of a factory etc., and the thing discharged from the mobile sources of a car etc., but especially the nitrogen oxides and soot dust from a diesel rolling stock pose a big problem triggered by the difficulty of the yield and purification.

[0003] In order to solve the aforementioned problem in a diesel rolling stock, engine improvement of the improvement of an air-supply-and-exhaust system by an improvement of a combustion chamber, an improvement of a fuel injection system, supercharge, etc., exhaust gas recirculation, etc. is considered, and nitrogen oxides and soot dust are being reduced. In order to solve an aforementioned problem, the improvement of fuel or a lubricating oil is also tried.

[0004] Development of the removal of an inflammable organic component (SOF) by an oxidation catalyst, prehension of the soot dust by a diesel particulate filter (DPF), a NOx reduction catalyst, etc. is furthered as post-processing art of exhaust gas.

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EFFECT OF THE INVENTION

[Effect of the Invention]This invention has the strong point in which high cleaning capacity can be further maintained over a long period of time compared with the former, without using special reducing agents, such as hydrocarbon, for example so that clearly from the place described above.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] However, in a diesel power plant, discharge control of nitrogen oxides and discharge control of soot dust have a relation of a trade-off, and there is a limit in those reduction effects only by engine improvement. The improvement of fuel or a lubricating oil also has many the problems and technical technical problems of cost.

[0006] in removal of SOF by the oxidation catalyst which is post-processing art, technical problems, such as improvement in low-temperature activity and control of sulfate generation at an elevated temperature, occur, and nitrogen oxides also have the problem that it cannot control. Although the crack initiation by the temperature gradient at the time of reproduction is a problem and DPF is being solved by improvement of a regeneration method, nitrogen oxides cannot be controlled in this method. For this reason, although development of the NOx reduction catalyst which operates under hyperoxia conditions like diesel-rolling-stock exhaust gas is furthered, it is still a research stage from points, such as shortage of cleaning capacity and the necessity for a reducing agent.

[0007] Thus, the conventional post-processing art had many problems, and it had the technical problem were difficult that any method removes nitrogen oxides and soot dust simultaneously substantially.

[0008] This invention can remove substantially simultaneously the soot dust and nitrogen oxides which are discharged from a combustion engine in view of such a technical problem of a Prior art, And it aims at providing the regeneration method of the stripper of soot dust and nitrogen oxides which can maintain high removal ability over a long period of time compared with the former more, and the removing method of soot dust and nitrogen oxides, without using special reducing agents, such as hydrocarbon.

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MEANS

[Means for Solving the Problem] This invention of claim 1 heats a filter which performs prehension of soot dust, and absorption of nitrogen oxides, It is a regeneration method of a stripper of soot dust and nitrogen oxides which reproduces said filter by burning soot dust caught by said filter, making predetermined reducing atmosphere using the combustion, and returning said absorbed nitrogen oxides.

[0010] This invention of claim 2 with a filter which has a prehension function of soot dust, and an oxidation absorption function of nitrogen oxides. While burning soot dust caught by filter by heating a filter and making the neighborhood of a filter into hypoxia atmosphere before it purifies soot dust and nitrogen oxides and absorptance or soot dust capturing capacity power of nitrogen oxides declines, After returning nitrogen oxides absorbed by filter and recovering absorptance of nitrogen oxides, it is a removing method of soot dust and nitrogen oxides which completes combustion of soot dust which remains in a filter, and reproduces a filter.

[0011] While this invention of claim 3 adds a steam at the time of filter regeneration, performs combustion of soot dust and steam reforming of soot dust which were caught by filter, for example and makes the neighborhood of a filter hypoxia atmosphere, After making nitrogen oxides absorbed by filter return and recovering absorptance of nitrogen oxides, it is a removing method of soot dust and nitrogen oxides which completes combustion of soot dust which remains in a filter, and reproduces a filter.

[0012] Furthermore, when an oxygen supply part is provided downstream from the above-mentioned filter, a catalyst which has an oxidation function is installed downstream from an oxygen supply part and an unburnt ingredient is discharged from a filter, this invention of claim 4, It is a removing method of soot dust and nitrogen oxides which introduces gas which contains oxygen from an oxygen supply part, and removes an unburnt ingredient according to an oxidation catalyst.

[0013] This invention of claim 5 is a removing method of soot dust and nitrogen oxides which uses for heating at the time of filter regeneration reaction fever generated in the above-mentioned oxidation catalyst.

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OPERATION

[Function] In this invention, the filter for ceramic monolithic wall style type particulates is mentioned, for example as an example of the filter used. This filter includes the catalyst which has a function which oxidizes nitric oxide to nitrogen dioxide under hyperoxia conditions, a function which absorbs nitrogen dioxide, and the function to return the nitrogen oxides which ****ed and were desorbed from the nitrogen oxides which the oxygen density absorbed in the atmosphere below a stoichiometric ratio to combustion of an unburnt ingredient further. As an example of a catalyst used by this invention, the catalyst which supported alkaline-earth metals, or a rare earth metal oxide and the precious metals to alumina is mentioned. Even nitrogen dioxide can oxidize nitric oxide by the oxidation of the bottom precious metals of a hyperoxia condition, and this catalyst can be made to absorb as a nitrate with metal, such as Ba and La, further. It can oxidize an unburnt ingredient while returning the nitrogen oxides which ****ed and were desorbed from the nitrogen oxides which were being absorbed to nitrogen, if this catalyst heats [an oxygen density] in the atmosphere below a stoichiometric ratio to combustion of an unburnt ingredient. It is thought that reduction of nitrogen oxides and combustion of an unburnt ingredient are the same mechanisms as a three way component catalyst. The multiple oxide containing Y, Ba, and Cu and the multiple oxide containing Mn and Zr may be used as a nitrogen oxide absorbent of this catalyst.

[0015] The regeneration method of the soot dust of this invention and the stripper of nitrogen oxides and the simultaneous removing method of soot dust and nitrogen oxides are explained collectively, describing hereafter the composition and operation of a purge which are shown in drawing 1.

[0016] Usually, exhaust gas is circulated in the filter 5 and removal of soot dust and removal of nitrogen oxides are performed by the above-mentioned principle. When constant value with the absorptance of the back pressure which increases by deposition of soot dust, or nitrogen oxides is exceeded, the valve 2 located in the filter 5 upper stream is switched, and the inflow of the exhaust gas to the filter 5 is suspended. On the other hand, air is supplied to the filter 5 with a pump, and the heater 4 performs heating of air and the filter 5. Although the combustion temperature of soot dust is usually 500-600 **, combustion at low temperature is attained by operation of the catalyst included in the filter 5. although the combustion temperature of the soot dust at the time of using a catalyst is based also on a controlled atmosphere and the description of soot dust -- 300-500 -- it becomes tens of **. If combustion of soot dust starts with heating, about five-filter oxygen will be consumed. Air supply is adjusted so that about five filter may become reducing atmosphere at this time. By this, desorption of the absorbed nitrogen oxides and reduction of the nitrogen oxides by partial oxidation output take place on a catalyst. When desorption of the absorbed

nitrogen oxides is fully performed, combustion of the increase of air supply and the soot dust which remains in the filter 5 without burning is completed. In this way, after reproducing the soot dust capturing capacity power of the filter 5, and the absorptance of nitrogen oxides, cleaning capacity is maintainable over a long period of time by purifying exhaust gas again.

[0017]Like a diesel rolling stock, when there are also many sulfur oxides in exhaust gas, a sulfur oxide is also absorbed by the catalyst, but. Since it will **** if the absorbed sulfur oxide is also exposed to an elevated temperature under reducing atmosphere, the fall of nitrogen-oxides absorptance can be controlled by burning the soot dust under reducing atmosphere at an elevated temperature further.

[0018]If a filter is installed in parallel and another filter purifies exhaust gas during filter regeneration of one of the two, purification of exhaust gas can be performed continuously substantially.

[0019]Although here described the method by an electric heater as the filter heating method, it is also possible to carry out direct heating of the soot dust with microwave.

[0020]With a strong substance of reducing power like [if a steam is added at the time of filter regeneration] the carbon monoxide which the oxygen tension near the filter could be reduced more easily and was further generated by steam reforming of soot dust, or hydrogen which burns easily. The conversion efficiency to nitrogen of the absorbed nitrogen oxides can be raised. In this case, it is desirable to make a filter contain the catalyst which promotes steam reforming like platinum or nickel. When desorption of the absorbed nitrogen oxides is completed, the operation which removes the soot dust which remained on the filter, and the carbon which deposited by steam reforming by combustion is the same as that of the method shown above.

[0021]Although the technical problem that it is generated by partial oxidation output occurs at the time of the reduction to nitrogen of the absorbed nitrogen oxides, This is solvable by providing an oxygen supply part downstream from a filter, installing the catalyst which has an oxidation function downstream from an oxygen supply part, introducing the gas which contains oxygen from an oxygen supply part, and oxidizing an unburnt ingredient according to an oxidation catalyst.

[0022]In the way an oxidation catalyst removes an unburnt ingredient, the reaction fever generated at the time of unburnt ingredient oxidation can be used for heating of a filter by installing an oxidation catalyst near the filter.

[Translation done.]

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EXAMPLE

[Example] Hereafter, the example of this invention is described, referring to drawings.

[0024] The regeneration method of the stripper of the soot dust and nitrogen oxides concerning other inventions is also explained simultaneously, describing one typical example of the removing method of soot dust and nitrogen oxides concerning this invention using drawing 1. Here, the stripper of soot dust and nitrogen oxides corresponds to the filter 5 of this example.

[0025] In drawing 1 -- 1 -- an exhaust gas introducing pipe and 2 -- as for a filter and 6, an air supply part and 4 are [a bypass and 8] exhaust gas discharge pipes a filter holding container and 7 a heater and 5 a valve and 3.

[0026] A below concrete example is explained in more detail.

[0027] A total of 300 g of catalysts which contain alumina, a lanthanum trioxide, barium oxide, cerium oxide, and the precious metals in the exhaust gas filter of the quality of mullite of the capacity of $1 > 3$ l. of < examples was supported. Making the lanthanum trioxide in a catalyst into 50wt%, the precious metals supported a total of Pt and 5 g/l of Rh(s).

[0028] This filter 5 was installed in the exhaust air part of a direct injection diesel engine with a displacement of 2400 cc, the valve 2 was formed in the upstream part of the filter 5, and the bypass 7 which can discharge exhaust gas as it is was installed. The heater 4 was formed in filter 5 front face, and the air supply part 3 was formed between the heater 4 and the valve 2. Space velocity was made into 60000h^{-1} , and emission gas was analyzed by the chemiluminescence type NOx analyzer, the hydrocarbon analyzer, and the gas chromatograph.

[0029] The presentations of filter 5 entrance gas at this time were about 500 ppm of nitric oxide, the sulfur dioxide of about 100 ppm, the amount of total hydrocarbon of about 500 ppm (methane conversion), about 4% of carbon dioxide, about 15% of oxygen, and about 10% of water.

[0030] Nitric oxide could carry out the extraction ratio about 90% at the time of an experiment start, and the extraction ratio fell with time. When the extraction ratio cut 30%, while passing the exhaust gas which circulates the filter 5 by the valve 2 to the bypass 7 and carrying out about 1 l/min supply of the air from the air supply part 3 further, heating of the heater 4 was started. When combustion of soot dust started, the discharged nitrogen oxide concentration became high temporarily, but it fell immediately. After the temperature of the filter 5 became about 650 ** and was fully desorbed from the sulfur compound, the amount of air supply which circulates the filter 5 was raised, and combustion of the soot dust accumulated in the filter 5 was completed. Usually, it was 33% when the mean clearance rate of nitrogen oxides was

calculated from the quantity of the discharge nitrogen oxides at the time of operation, and the quantity of the nitrogen oxides discharged at the time of filter 5 reproduction.

[0031]Although air was used for reproduction here, it is also possible to use a part of exhaust gas which is a hypoxia partial pressure as resurgent gas. In this case, more efficient reproduction is possible if the combustion temperature of the exhaust gas at the time of reproduction is raised by throttling using an inhalation-of-air diaphragm or an exhaust air diaphragm.

[0032]The catalyst of the filter 5 used in <Example 2> example 1 was made to contain nickel oxide. It was made for the weight of nickel oxide to be 10wt% into a catalyst.

[0033]First, like Example 1, perform absorption of nitrogen oxides, and prehension of soot dust, and the air which contained 50vol% of the steam from the air supply part 3 at the time of reproduction is circulated by about 1 l/min, When the temperature of the filter 5 became 650 **, supply of the steam was suspended, only air was circulated, and combustion of soot dust was completed like Example 1. On this condition, from the quantity of the discharge nitrogen oxides at the time of operation, and the quantity of the nitrogen oxides by which it was generated at the time of filter 5 reproduction, when the mean clearance rate of nitrogen oxides was searched for, compared with the case of 38% and Example 1, the nitrogen-oxides extraction ratio usually improved.

[0034]From the above result, by adding a steam in a filter at the time of reproduction showed that the reduction to nitrogen of the absorbed nitrogen oxides was promoted.

[0035]A total of 100 g of catalysts which contain alumina, Pt and Rh, and cerium oxide in the honeycomb of the cordierite of the capacity of 3> 1 l. of < examples was made to support, and the oxidation catalyst 16 was prepared.

[0036]As shown in (drawing 2), the air supply part 15 for unburnt ingredient purification was formed downstream from the filter 13, the oxidation catalyst 16 was installed downstream from the air supply part 15 for unburnt ingredient purification, and the heater 17 which heats the oxidation catalyst 16 was formed.

[0037]First, absorption of nitrogen oxides and prehension of soot dust were performed like Example 1. After that, at the time of reproduction, air was supplied by 1 l/min from the air supply part 15 for unburnt ingredient purification, and the oxidation catalyst 16 was heated at 450 ** with the heater 17. Reproduction was performed by the same method as Example 1, when reproduction was completed, it suspended introduction of the air from the air supply part 15 for unburnt ingredient purification, and heating of the oxidation catalyst 16 also suspended it. When the hydrocarbon concentration of the lower stream of the oxidation catalyst 16 was measured by the hydrocarbon analyzer and it compared with the case of Example 1 at the time of reproduction, it became 17% when the amount of total hydrocarbon discharged at the time of reproduction is not installing the oxidation catalyst 16, and was decreasing greatly.

[0038]As a result, it turned out that the quantity of the unburnt ingredient generated at the time of reproduction can be greatly reduced according to an oxidation catalyst.

[0039]The ***** cordierite honeycomb was created for the central part in the form surrounding [as shown in <Example 4> (drawing 3)] the periphery of the filter holding container 25. Honeycomb capacity was 1 l. By supporting a catalyst like Example 3 to this honeycomb, the oxidation catalyst 27 was prepared and the filter holding container 25 was installed with the wrap form.

[0040]On the other hand, the emission gas from the filter 24 changed the channel so that the oxidation catalyst 27 might be passed, and it formed the air supply part 26 for unburnt ingredient purification between

the filter 24 and the oxidation catalyst 27. The heater 28 was formed so that the periphery of an oxidation catalyst holding container could be heated.

[0041]First, absorption of nitrogen oxides and prehension of soot dust were performed like Example 1. After that, at the time of reproduction, air was supplied by 1 l/min from the air supply part 26 for unburnt ingredient purification, and the oxidation catalyst 27 was heated at 450 **. Reproduction of the filter 24 was performed by the same method as Example 1, when reproduction was completed, it suspended introduction of the air of the air supply part 26 for unburnt ingredient purification, and heating of the oxidation catalyst 27 also suspended it. When the time of the time of the end of desorption of a sulfur compound was measured in the meantime, it turned out that time is shortened by 5 by about 3/in the case of Example 1.

[0042]From the above result, by installing this oxidation catalyst near the filter showed that reaction fever could be used for heating of a filter and a filter could be reproduced efficiently by the way an oxidation catalyst removes an unburnt ingredient.

[0043]Although the above-mentioned example explained the case where a filter was reproduced by completing combustion of the soot dust which reproduced the absorptance of the nitrogen oxides of a filter and was caught by the filter after that by returning the nitrogen oxides absorbed by the filter 5, If the capturing capacity power of the soot dust of a filter is recovered to some extent by combustion of soot dust performed under hypoxia atmosphere when returning the nitrogen oxides absorbed by not only this but the filter, after reproducing the absorptance of nitrogen oxides, it is not necessary to perform combustion of the still more above soot dust.

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is a figure showing the section of the example of the purge used by this invention.

[Drawing 2] It is the figure which was used by this invention and in which showing the section of the example of the purge provided with the oxidation catalyst.

[Drawing 3] It is the figure which was used by this invention and in which showing the section of the example of the purge which uses for filter regeneration the heat generated in an oxidation catalyst.

[Description of Notations]

- 1 Exhaust gas introducing pipe
- 2 Valve
- 3 Air supply part
- 4 Heater
- 5 Filter
- 6 Filter holding container
- 7 Bypass
- 8 Exhaust gas discharge pipe
- 9 Exhaust gas introducing pipe
- 10 Valve
- 11 Air supply part
- 12 Heater
- 13 Filter
- 14 Filter holding container
- 15 The air supply part for unburnt ingredient purification
- 16 Oxidation catalyst
- 17 Heater
- 18 Bypass
- 19 Exhaust gas discharge pipe
- 20 Exhaust gas introducing pipe
- 21 Valve
- 22 Air supply part
- 23 Heater

- 24 Filter
 - 25 Filter holding container
 - 26 The air supply part for unburnt ingredient purification
 - 27 Oxidation catalyst
 - 28 Heater
 - 29 Bypass
 - 30 Exhaust gas discharge pipe
-

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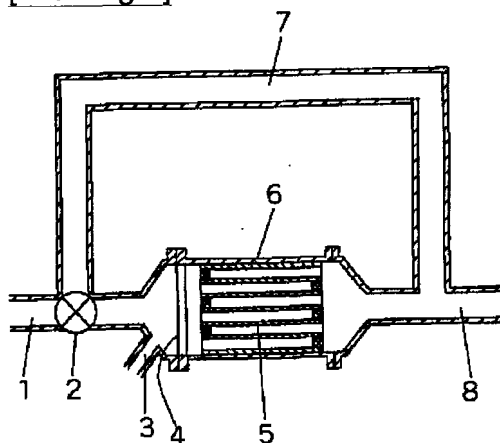
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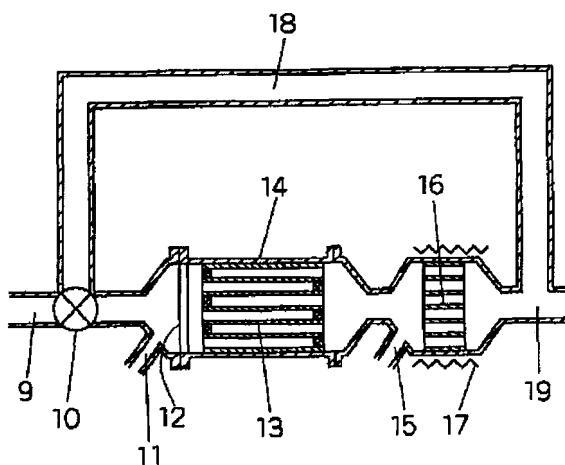
DRAWINGS

[Drawing 1]



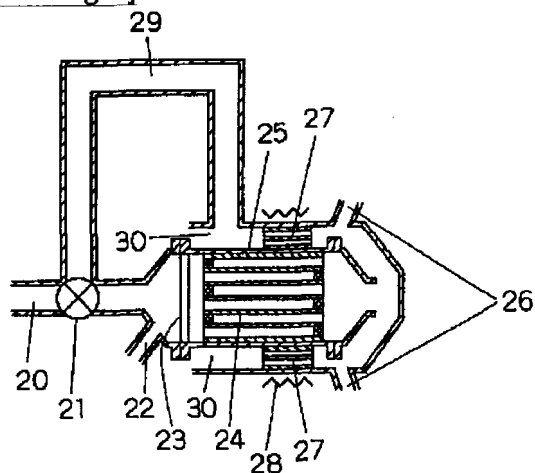
- 1 : 排気ガス導入管
- 2 : バルブ
- 3 : 空気供給部
- 4 : ヒータ
- 5 : フィルタ
- 6 : フィルタ保持容器
- 7 : バイパス
- 8 : 排気ガス排出管

[Drawing 2]



- 9 : 排気ガス導入管
 10 : バルブ
 11 : 空気供給部
 12, 17 : ヒータ
 13 : フィルタ
 14 : フィルタ保持容器
 15 : 未燃成分浄化用空気供給部
 16 : 酸化触媒
 18 : バイパス
 19 : 排気ガス排出管

[Drawing 3]



- 20 : 排気ガス導入管
 21 : バルブ
 22 : 空気供給部
 23, 28 : ヒータ
 24 : フィルタ
 25 : フィルタ保持容器
 26 : 未燃成分浄化用空気供給部
 27 : 酸化触媒
 29 : バイパス
 30 : 排気ガス排出管

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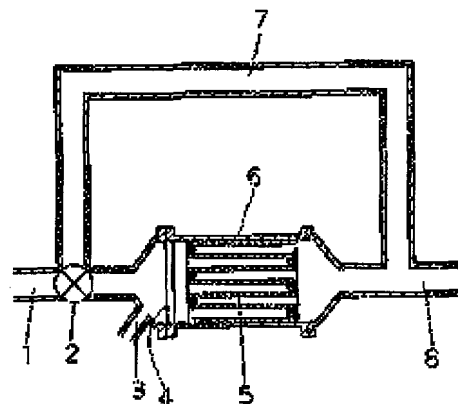
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(54) 【発明の名称】 煤塵と窒素酸化物の除去装置の再生方法、及び煤塵と窒素酸化物の除去方法

(57) 【要約】

【目的】 ディーゼル機関などから排出される煤塵と窒素酸化物を同時に除去する方法に関するもので、炭化水素などの特殊な還元剤を用いることなく、長期にわたり高い能力を維持できる浄化方法を提供することを目的としている。

【構成】 煤塵の捕捉機能と窒素酸化物の酸化吸収機能を有するフィルタ5により煤塵と窒素酸化物を浄化し、窒素酸化物の吸収能力が飽和に達する前にフィルタ5を加熱することによりフィルタ5に捕捉された煤塵を燃焼させフィルタ5近傍を低酸素雰囲気とするとともに、フィルタ5に吸収された窒素酸化物を還元し、窒素酸化物の吸収能力を再生する。その後、煤塵の燃焼を完結させフィルタ5を再生する。これを繰り返すことにより、長期にわたりディーゼル機関からの排出物を浄化し得る。



1: 排気ガス導入管

7: バルブ

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【特許請求の範囲】

【請求項1】 煤塵の捕捉と窒素酸化物の吸収を行なうフィルタを加熱して、前記フィルタに捕捉された煤塵を燃焼させ、その燃焼を利用して所定の還元雰囲気を作り出し、前記吸収された窒素酸化物を還元することにより、前記フィルタを再生させることを特徴とする煤塵と窒素酸化物の除去装置の再生方法。

【請求項2】 煤塵の捕捉機能と窒素酸化物の酸化吸収機能を有するフィルタにより、煤塵と窒素酸化物を浄化し、窒素酸化物の吸収能力もしくは煤塵の捕捉能力が低下する前に前記フィルタを加熱することにより前記フィルタに捕捉された煤塵を燃焼させ前記フィルタ近傍を低酸素雰囲気にするとともに、前記フィルタに吸収された窒素酸化物を還元し、窒素酸化物の吸収能力を回復させた後、前記フィルタに残留する煤塵の燃焼を完結させ前記フィルタを再生することを特徴とする煤塵と窒素酸化物の除去方法。

【請求項3】 煤塵の捕捉機能と窒素酸化物の酸化吸収機能を有するフィルタにより、煤塵と窒素酸化物を浄化し、窒素酸化物の吸収能力もしくは煤塵の捕捉能力が低下する前に前記フィルタを加熱し、かつ水蒸気を加えることにより前記フィルタに捕捉された煤塵の燃焼および煤塵の水蒸気改質を行い前記フィルタ近傍を低酸素雰囲気にするとともに、前記フィルタに吸収された窒素酸化物を還元し、窒素酸化物の吸収能力を回復させた後、前記フィルタに残留する煤塵の燃焼を完結させ前記フィルタを再生することを特徴とする煤塵と窒素酸化物の除去方法。

【請求項4】 フィルタの下流に酸素供給部を設け、前記酸素供給部の下流に酸化機能を有する触媒を設置し、前記フィルタから未燃成分が排出された時点で、前記酸素供給部から酸素を含むガスを導入し、前記酸化触媒により前記未燃成分を除去することを特徴とする請求項2または3記載の煤塵と窒素酸化物の除去方法。

【請求項5】 酸化触媒で発生する反応熱をフィルタ再生時の加熱に利用することを特徴とする請求項4に記載の煤塵と窒素酸化物の除去方法。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は、例えば、ディーゼル機関等から排出される煤塵と窒素酸化物を同時に除去する場合等に利用可能な、煤塵と窒素酸化物の除去装置の再生方法、及び煤塵と窒素酸化物の除去方法に関するものである。

【0002】

【従来の技術】近年、大都市圏における窒素酸化物や煤塵による大気汚染が問題となっており、早急な対策が望まれている。これらは、大きく分けると工場などの固定発生源から排出されるものと、自動車などの移動発生源から排出されるものに分けられるが、特にディーゼル車

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からの窒素酸化物と煤塵はその発生量と浄化の困難さから、大きな問題となっている。

【0003】ディーゼル車における上記課題を解決するために、燃焼室の改善、燃料噴射系の改善、過給などによる給排気系の改善、排気ガス再循環などのエンジン改良が検討されており、窒素酸化物と煤塵は低減されつつある。また、上記課題を解決するため燃料や潤滑油の改善も試みられている。

【0004】さらに、排気ガスの後処理技術として、酸化触媒による可燃性有機成分(SOF)の除去や、ディーゼルパティキュレートフィルター(DPF)による煤塵の捕捉、NOx還元触媒などの開発が進められている。

【0005】

【発明が解決しようとする課題】しかし、ディーゼルエンジンにおいては、窒素酸化物の排出抑制と煤塵の排出抑制はトレードオフの関係にあり、エンジンの改良だけではそれらの低減効果には限界がある。また燃料や潤滑油の改善もコストの問題や技術的課題が多い。

【0006】さらに、後処理技術である酸化触媒によるSOFの除去では、低温活性の向上と高温でのサルフェート生成の抑制等の課題があり、また窒素酸化物は抑制できないといった問題もある。また、DPFは再生時の温度勾配による亀裂の発生が問題であり、再生方法の改善により解決されつつあるものの、本方法においても窒素酸化物は抑制できない。このためディーゼル車排気のような酸素過剰条件下で作動するNOx還元触媒の開発が進められているが、浄化能力の不足、還元剤の必要性などの点から未だ研究段階である。

【0007】このように、従来の後処理技術は、多くの問題を抱えていると共に、いずれの方法も窒素酸化物と煤塵を実質的に同時に除去することは困難であるといった課題を有していた。

【0008】本発明は、従来の技術のこのような課題に鑑み、燃焼機関から排出される煤塵と窒素酸化物を実質的に同時に除去出来、しかも、炭化水素などの特殊な還元剤を用いることなく、従来に比べてより長期にわたり高い除去能力を維持出来る煤塵と窒素酸化物の除去装置の再生方法、及び煤塵と窒素酸化物の除去方法を提供することを目的としている。

【0009】

【課題を解決するための手段】請求項1の本発明は、煤塵の捕捉と窒素酸化物の吸収を行なうフィルタを加熱して、前記フィルタに捕捉された煤塵を燃焼させ、その燃焼を利用して所定の還元雰囲気を作り出し、前記吸収された窒素酸化物を還元することにより、前記フィルタを再生させる煤塵と窒素酸化物の除去装置の再生方法である。

【0010】請求項2の本発明は、煤塵の捕捉機能と窒素酸化物の酸化吸収機能を有するフィルタにより、煤塵

と窒素酸化物を浄化し、窒素酸化物の吸収能力もしくは煤塵捕捉能力が低下する前にフィルタを加熱することによりフィルタに捕捉された煤塵を燃焼させフィルタ近傍を低酸素雰囲気にするるとともに、フィルタに吸収された窒素酸化物を還元し、窒素酸化物の吸収能力を回復させた後、フィルタに残留する煤塵の燃焼を完結させフィルタを再生する煤塵と窒素酸化物の除去方法である。

【0011】また請求項3の本発明は、例えば、フィルタ再生時に水蒸気を加え、フィルタに捕捉された煤塵の燃焼および煤塵の水蒸気改質を行いフィルタ近傍を低酸素雰囲気にするるとともに、フィルタに吸収された窒素酸化物を還元させ、窒素酸化物の吸収能力を回復させた後、フィルタに残留する煤塵の燃焼を完結させフィルタを再生する煤塵と窒素酸化物の除去方法である。

【0012】さらに請求項4の本発明は、上記フィルタの下流に酸素供給部を設け、酸素供給部の下流に酸化機能を有する触媒を設置し、フィルタから未燃成分が排出された時点で、酸素供給部から酸素を含むガスを導入し、酸化触媒により未燃成分を除去する煤塵と窒素酸化物の除去方法である。

【0013】請求項5の本発明は、上記酸化触媒で発生する反応熱をフィルタ再生時の加熱に利用する煤塵と窒素酸化物の除去方法である。

【0014】

【作用】本発明では、例えば、用いられるフィルタの例として、セラミックモノリシック壁流型微粒子物用フィルタが挙げられる。本フィルタは、酸素過剰条件下で一酸化窒素を二酸化窒素に酸化する機能と二酸化窒素を吸収する機能と、さらに酸素濃度が未燃成分の燃焼に対して化学量論比以下の雰囲気中で吸収した窒素酸化物を脱離し、かつ脱離した窒素酸化物を還元する機能を有する触媒を含む。本発明で用いられる触媒の一例として、アルカリ土類金属もしくは希土類金属酸化物と貴金属をアルミナに担持した触媒が挙げられる。この触媒は、酸素過剰条件下貴金属の酸化作用により一酸化窒素を二酸化窒素まで酸化し、さらにBa、Laなどの金属により硝酸塩として吸収させることができる。また本触媒は酸素濃度が未燃成分の燃焼に対して化学量論比以下の雰囲気中で加熱すると、吸収していた窒素酸化物を脱離し、かつ脱離した窒素酸化物を窒素に還元するとともに、未燃成分を酸化することができる。窒素酸化物の還元と未燃成分の燃焼は三元触媒と同様の機構であると考えられている。なお、本触媒の窒素酸化物吸収剤として、Y、Ba、Cuを含む複合酸化物やMn、Zrを含む複合酸化物を用いてもよい。

【0015】以下、図1に示す浄化装置の構成及び動作を述べながら、本発明の煤塵と窒素酸化物の除去装置の再生方法、及び煤塵と窒素酸化物の同時除去方法についてまとめて説明する。

【0016】通常は、フィルタ5に排気ガスを流通さ

せ、煤塵の除去と窒素酸化物の除去を上記原理で行う。煤塵の堆積により増加する背圧もしくは窒素酸化物の吸収能力がある一定値を越えた時点で、フィルタ5上流に位置するバルブ2を切り換え、フィルタ5への排気ガスの流入を停止する。一方、フィルタ5にはポンプにより空気を供給し、ヒータ4により空気とフィルタ5の加熱を行う。煤塵の燃焼温度は通常500～600℃であるが、フィルタ5中に含まれる触媒の作用により低温での燃焼が可能となる。触媒を用いた場合の煤塵の燃焼温度は、雰囲気ガスや、煤塵の性状にもよるが、300～500数十℃となる。加熱により煤塵の燃焼が始まると、フィルタ5近傍の酸素が消費される。この時点でフィルタ5近傍が還元雰囲気になるように空気供給量を調節する。これによって、吸収された窒素酸化物の脱離と部分酸化生成物による窒素酸化物の還元が触媒上で起こる。吸収された窒素酸化物の脱離が十分に行われた時点で、空気供給量を増し、燃焼されずにフィルタ5に残留している煤塵の燃焼を完結させる。こうして、フィルタ5の煤塵捕捉能力と窒素酸化物の吸収能力を再生させた後、再び排気ガスの浄化を行うことにより、長期にわたり浄化能力を維持することができる。

【0017】なお、ディーゼル車のように排気ガス中の硫黄酸化物も多い場合は、硫黄酸化物も触媒に吸収されるが、吸収された硫黄酸化物も還元雰囲気下で高温にさらすと脱離するため、還元雰囲気下の煤塵の燃焼をさらに高温で行うことにより、窒素酸化物吸収能力の低下を抑制することができる。

【0018】また、フィルタを並列に設置し片方のフィルタ再生中にもう一方のフィルタで排気ガスの浄化を行えば、実質的に連続して排気ガスの浄化ができる。

【0019】フィルタ加熱方法として、ここでは電気ヒータによる方法について述べたが、マイクロ波で煤塵を直接加熱することも可能である。

【0020】また、フィルタ再生時に水蒸気を加えれば、より容易にフィルタ近傍の酸素分圧を低下させることができ、さらに煤塵の水蒸気改質により生成した一酸化炭素や水素のような燃焼し易く還元力の強い物質により、吸収された窒素酸化物の窒素への変換効率を向上させることができる。この場合には、白金やニッケルのような水蒸気改質を促進させる触媒をフィルタに含有させることが望ましい。なお、吸収された窒素酸化物の脱離が完了した時点で、フィルタ上に残った煤塵と水蒸気改質により析出した炭素を燃焼により取り除く操作は上記で示した方法と同様である。

【0021】さらに、吸収された窒素酸化物の窒素への還元時に、部分酸化生成物が発生するといった課題があるが、フィルタの下流に酸素供給部を設け、酸素供給部の下流に酸化機能を有する触媒を設置し、酸素供給部から酸素を含むガスを導入し、酸化触媒により未燃成分を酸化することによりこれを解決することができる。

【0022】また、酸化触媒により未燃成分を除去する方法では、酸化触媒をフィルタ近傍に設置することにより、未燃成分酸化時に発生する反応熱をフィルタの加熱に利用することができる。

【0023】

【実施例】以下、本発明の実施例を図面を参照しながら説明する。

【0024】本発明にかかる煤塵と窒素酸化物の除去方法の代表的な一実施例を、図1を利用して述べながら、他の発明にかかる煤塵と窒素酸化物の除去装置の再生方法について同時に説明する。ここで、煤塵と窒素酸化物の除去装置は、本実施例のフィルタ5に対応する。

【0025】図1において、1は排気ガス導入管、2はバルブ、3は空気供給部、4はヒータ、5はフィルタ、6はフィルタ保持容器、7はバイパス、8は排気ガス排出管である。

【0026】以下に具体的な例について、更に詳しく説明する。

【0027】＜実施例1＞3リットルの容積のムライト質の排ガスフィルタにアルミナと酸化ランタンと酸化バリウムと酸化セリウムおよび貴金属を含む触媒計300gを担持した。触媒中の酸化ランタンは50wt%とし、貴金属はPtとRhを計5g/1担持した。

【0028】このフィルタ5を排気量2400ccの直接噴射式ディーゼルエンジンの排気部に設置し、フィルタ5の上流部にバルブ2を設け、排気ガスをそのまま排出できるバイパス7を設置した。さらに、フィルタ5前面にヒータ4を設け、ヒータ4とバルブ2の間に空気供給部3を設けた。なお、空間速度は6000h⁻¹とし、排出ガスは化学発光式NOx分析計、炭化水素計、ガスクロマトグラフで分析した。

【0029】このときの、フィルタ5入り口ガスの組成は、一酸化窒素約500ppm、二酸化硫黄約100ppm、全炭化水素量（メタン換算）約500ppm、二酸化炭素約4%、酸素約15%、水約10%であった。

【0030】一酸化窒素は、実験開始時には90%程度除去率で、時間と共に除去率は低下した。除去率が30%をきった時点で、バルブ2によりフィルタ5に流通させる排ガスをバイパス7に流し、さらに空気供給部3より空気を約1l/min供給するとともに、ヒータ4の加熱を開始した。煤塵の燃焼が始まった時点で、排出された窒素酸化物濃度が一時的に高くなったが、すぐに低下した。フィルタ5の温度が約650℃になり、十分に炭素化合物の脱離を行った後フィルタ5に流通させる空気の供給量を上げ、フィルタ5に蓄積した煤塵の燃焼を完結させた。通常運転時の排出窒素酸化物の量とフィルタ5再生時に排出された窒素酸化物の量から、窒素酸化物の平均除去率を求めたところ33%であった。

【0031】なお、ここでは再生に空気をを用いたが、低酸素分圧である排気ガスの一部を再生ガスとして用いる

ことも可能である。この場合には、吸気絞りまたは排気絞りをを用いるスロットリングにより、再生時の排気ガスの燃焼温度をあげれば、より効率的な再生が可能である。

【0032】＜実施例2＞実施例1で用いたフィルタ5の触媒に酸化ニッケルを含有させた。酸化ニッケルの重量は触媒中に10wt%となるようにした。

【0033】まず、実施例1と同様に、窒素酸化物の吸収と煤塵の捕捉を行い、再生時に空気供給部3より50vol%の水蒸気を含んだ空気を約1l/minで流通させ、フィルタ5の温度が650℃になった時点で水蒸気の供給を停止し空気のみを流通させ、実施例1と同様に煤塵の燃焼を完結させた。この条件で、通常運転時の排出窒素酸化物の量とフィルタ5再生時に発生した窒素酸化物の量から、窒素酸化物の平均除去率を求めたところ38%と実施例1の場合に比べ窒素酸化物除去率が向上した。

【0034】以上の結果より、再生時にフィルタに水蒸気を添加することにより、吸収された窒素酸化物の窒素への還元が促進されることがわかった。

【0035】＜実施例3＞1リットルの容積のコージュライトのハニカムにアルミナとPtとRhおよび酸化セリウムを含む触媒計100gを担持させ、酸化触媒16を調製した。

【0036】（図2）に示すように、フィルタ13の下流に未燃成分浄化用空気供給部15を設け、未燃成分浄化用空気供給部15の下流に酸化触媒16を設置し、酸化触媒16を加熱するヒータ17を設けた。

【0037】まず、実施例1と同様に、窒素酸化物の吸収と煤塵の捕捉を行った。その後再生時には、未燃成分浄化用空気供給部15から空気を1l/minで供給し、ヒータ17により酸化触媒16を450℃に加熱した。再生は、実施例1と同様の方法で行い、再生が終了した時点で未燃成分浄化用空気供給部15からの空気の導入を停止し、酸化触媒16の加熱も停止した。なお、再生時に、酸化触媒16の下流の炭化水素濃度を炭化水素計で測定し、実施例1の場合と比較したところ、再生時に排出される全炭化水素量が酸化触媒16を設置していない場合の17%となり、大きく減少していた。

【0038】この結果、酸化触媒により、再生時に発生する未燃成分の量を大きく低減できることがわかった。

【0039】＜実施例4＞（図3）に示すようにフィルタ保持容器25の外周を囲む形に中心部をくり貫いたコージュライトハニカムを作成した。なお、ハニカム容積は1リットルとした。このハニカムに実施例3と同様に触媒を担持することにより、酸化触媒27を調製し、フィルタ保持容器25を覆う形で設置した。

【0040】一方、フィルタ24からの排出ガスが、酸化触媒27を通過するように流路を変更し、フィルタ24と酸化触媒27の間に未燃成分浄化用空気供給部26

を設けた。さらに、酸化触媒保持容器の外周を加熱できるようにヒータ28を設けた。

【0041】まず、実施例1と同様に、窒素酸化物の吸収と煤塵の捕捉を行った。その後再生時に、未燃成分浄化用空気供給部26から空気を1 l/minで供給し、酸化触媒27を450℃に加熱した。フィルタ24の再生は、実施例1と同様の方法で行い、再生が終了した時点で未燃成分浄化用空気供給部26の空気の導入を停止し、酸化触媒27の加熱も停止した。この間、硫黄化合物の脱着終了時までの時間を測定したところ、実施例1の場合の約3/5に時間が短縮されていることがわかった。

【0042】以上の結果より、酸化触媒により未燃成分を除去する方法では、本酸化触媒をフィルタ近傍に設置することにより、反応熱をフィルタの加熱に利用することができ、効率的にフィルタの再生を行うことができることがわかった。

【0043】尚、上記実施例では、フィルタ5に吸収された窒素酸化物を還元することによって、フィルタの窒素酸化物の吸収能力を再生しその後、そのフィルタに捕捉された煤塵の燃焼を完結させることにより、フィルタを再生させる場合について説明したが、これに限らず例えば、フィルタに吸収された窒素酸化物を還元する際に、低酸素雰囲気のもとで行われる煤塵の燃焼によって、フィルタの煤塵の捕捉能力がある程度回復しておれば、窒素酸化物の吸収能力を再生した後、更に上記のような煤塵の燃焼を行う必要はない。

【0044】

【発明の効果】以上述べたところから明らかなように本発明は、例えば炭化水素などの特殊な還元剤を用いることなく、従来に比べてより一層長期にわたり高い浄化能力を維持することができるという長所を有する。

【図面の簡単な説明】

【図1】本発明で用いた浄化装置の実施例の断面を示す図である。

【図2】本発明で用いた、酸化触媒を備えた浄化装置の

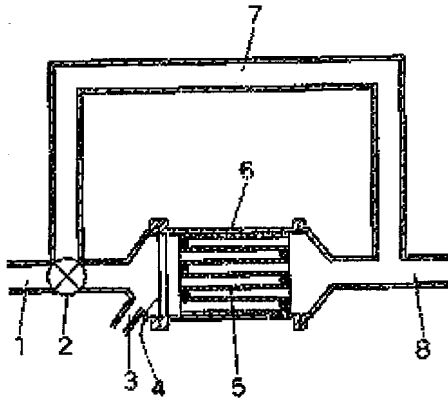
実施例の断面を示す図である。

【図3】本発明で用いた、酸化触媒で発生する熱をフィルタ再生に利用する浄化装置の実施例の断面を示す図である。

【符号の説明】

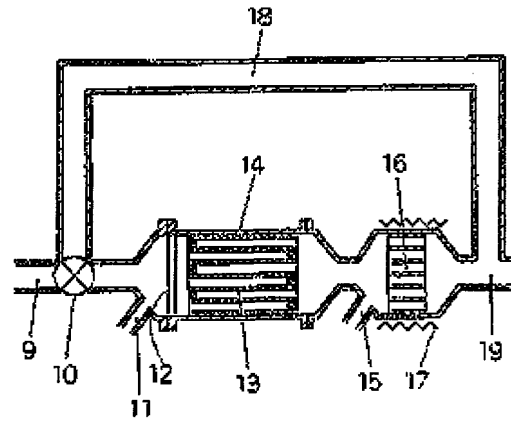
- | | |
|----|--------------|
| 1 | 排気ガス導入管 |
| 2 | バルブ |
| 3 | 空気供給部 |
| 4 | ヒータ |
| 5 | フィルタ |
| 6 | フィルタ保持容器 |
| 7 | バイパス |
| 8 | 排気ガス排出管 |
| 9 | 排気ガス導入管 |
| 10 | バルブ |
| 11 | 空気供給部 |
| 12 | ヒータ |
| 13 | フィルタ |
| 14 | フィルタ保持容器 |
| 15 | 未燃成分浄化用空気供給部 |
| 16 | 酸化触媒 |
| 17 | ヒータ |
| 18 | バイパス |
| 19 | 排気ガス排出管 |
| 20 | 排気ガス導入管 |
| 21 | バルブ |
| 22 | 空気供給部 |
| 23 | ヒータ |
| 24 | フィルタ |
| 25 | フィルタ保持容器 |
| 26 | 未燃成分浄化用空気供給部 |
| 27 | 酸化触媒 |
| 28 | ヒータ |
| 29 | バイパス |
| 30 | 排気ガス排出管 |

【図1】



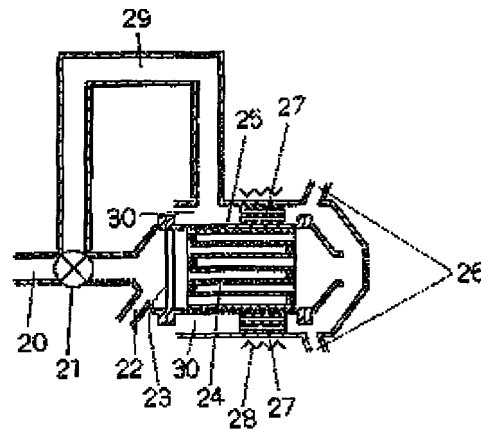
- 1: 排気ガス導入管
- 2: バルブ
- 3: 空気供給部
- 4: ヒータ
- 5: フィルタ
- 6: フィルタ保持容器
- 7: バイパス
- 8: 排気ガス排出管

【図2】



- 9: 排気ガス導入管
- 10: バルブ
- 11: 空気供給部
- 12, 17: ヒータ
- 13: フィルタ
- 14: フィルタ保持容器
- 15: 未燃成分浄化用空気供給部
- 16: 酸化触媒
- 18: バイパス
- 19: 排気ガス排出管

【図3】



- 20: 排気ガス導入管
 21: バルブ
 22: 空気供給部
 23, 28: ヒータ
 24: フィルタ
 25: フィルタ保持容器
 26: 末端成分浄化用室気供給部
 27: 酸化触媒
 29: バイパス
 30: 排気ガス集出管

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